

## PATENT

Docket No. 043887-0192

## IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

APPLICANTS: Takehiko YAMASHITA et al. CONF.: 9389  
APPLN. NO.: 10/572,857 ART UNIT: 1796  
FILED: March 22, 2006 EXAMINER: Gregory LISTVOYE  
FOR: FLAME-RETARDANT RESIN COMPOSITION, PRODUCTION  
METHOD OF THE SAME AND MOLDING METHOD OF THE  
SAME

***DECLARATION UNDER 37 C.F.R. § 1.132***

Assistant Commissioner of Patents  
Alexandria, VA 22313-1450

Sir:

I, Takehiko YAMASHITA, do declare and say as follows:

1. I am a graduate of the Department of Applied Chemistry of Faculty of Engineering, of Nagoya Institute of Technology, and received my bachelor's degree.

2. I am presently employed by MATSUSHITA ELECTRIC INDUSTRIAL CO., LTD. since March, 6, 1989.

3. I am listed as one of the inventors of the subject of the above-identified application, and I have read and understand the application.

4. I have read and understood the references cited in the Office Action notified on May 27, 2008.

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5. That the following experiments were carried out by me or under my direct supervision and control.

6. Experiments

The experiments were carried out as follows:

Experiment 1

Poly(lactic acid) (PLA) of 70wt%, which was synthesized from corn as a material and poly(butylene succinate) (PBS) of 30wt% were kneaded with a twin screw kneader and pellets were produced. Herein, PBS was added for the purpose of improving heat resistance.

In this experiment, acetylacetonatoiron ( $\text{Fe}(\text{acac})_3$ ) as a flame-retardant component was simply mixed with an  $\text{SiO}_2$  porous material. The pellets of 90wt% and the mixture of  $\text{Fe}(\text{acac})_3$  and  $\text{SiO}_2$  porous material of 10wt% were kneaded with the twin screw kneader at 185°C and press-molded into a test piece of 125mm x 13mm x 3.2mm (at a molding temperature of 180°C under a pressure of 120kg/cm<sup>2</sup>). The  $\text{SiO}_2$  porous material used in this experiment had a porosity of about 45vol% to about 50vol%, and a granular diameter of about 100nm to about 1000nm. This  $\text{SiO}_2$  porous material was crushed by a shearing force when being kneaded with the resin, and finally dispersed as finer particles which had a particle diameter of about 25nm to about 150nm (a mean particle diameter of about 75nm) in the resin.  $\text{Fe}(\text{acac})_3$  was used in a form of powder which had a particle diameter of about 2 $\mu\text{m}$  to about 80 $\mu\text{m}$ . The  $\text{Fe}(\text{acac})_3$

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powder was not crushed by kneading and the powder remaining the initial size was dispersed in the resin. The content of  $\text{Fe}(\text{acac})_3$  in the resin composition was calculated to be 3.75wt%.

This test piece was subjected to a 20mm vertical flame test according to Underwriters Laboratories-94. The result is shown in Table 1. From the test results, this sample was evaluated to be V2 according to the UL specification.

Experiment 2

Pellets were produced by kneading polylactic acid (PLA) and polybutylene succinate (PBS) following the same procedures as those in Experiment 1.

A blending sequence (order) for the composition in this experiment was similar to Experiment 1. In this experiment, zinc borate as the flame-retardant component was mixed with the  $\text{SiO}_2$  porous material. Zinc borate of 42 parts by weight was mixed with the porous material of 100 parts by weight. The pellets of 90wt% and the mixture of zinc borate and the  $\text{SiO}_2$  porous material of 10wt% were mixed with the twin screw kneader at 185°C, and press-molded into a test piece of 125mm x 13mm x 3.2mm (at a molding temperature of 180°C under a pressure of 120kg/cm<sup>2</sup>). The  $\text{SiO}_2$  porous material employed in this experiment was the same as that employed in Experiment 1 and dispersed in the resin in a form of fine particles in the order of nanometer by kneading. Zinc borate

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was used in a form of powder which had a particle diameter of about 5 $\mu$ m to about 100 $\mu$ m. The powder was not crushed by kneading and the powder remaining the initial size was dispersed in the resin. The content of zinc borate in the resin composition was calculated to be 3.0wt%.

The resultant test piece was subjected to the UL-94 vertical flame test similarly to Experiment 1. The results are shown in Table 1. From the results shown in Table 1, this sample was evaluated to be V2 according to the UL specification.

**Experiment 3**

Pellets were produced by kneading polylactic acid (PLA) and polybutylene succinate (PBS) following the same procedures as those in Experiment 1. A blending sequence (order) for the composition in this experiment was similar to Experiment 1. In this experiment, TBBA (tetrabromobisphenol-A) as the flame-retardant component was mixed with the SiO<sub>2</sub> porous particles. TBBA of 20 parts by weight was mixed with the porous particles of 100 parts by weight. The pellets of 90wt% and the mixture of TBBA and the SiO<sub>2</sub> porous particles of 10wt% were kneaded with the twin screw kneader at 185°C, and press-molded into a test piece of 125mm x 13mm x 3.2mm (at a molding temperature of 180°C under a pressure of 120kg/cm<sup>2</sup>). The SiO<sub>2</sub> porous particles employed in this experiment were the same as those employed in Experiment 1 and dispersed in the resin in a form of fine particles in the order of nanometer by kneading. TBBA

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was used in a form of powder which had a particle diameter of about 20 $\mu$ m to about 100 $\mu$ m. The powder was not crushed by kneading and the powder remaining the initial size was dispersed in the resin. The content of TBBA in the resin composition was calculated to be 1.7wt%.

The resultant test piece was subjected to the UL-94 vertical flame test similarly to Experiment 1. The results are shown in Table 1. From the results shown in Table 1, this sample was evaluated to be V2 according to the UL specification.

Experiment 4

Pellets were produced by kneading polylactic acid (PLA) and polybutylene succinate (PBS) following the same procedures as those in Experiment 1. A blending sequence (order) for the composition in this experiment was similar to Experiment 1. In this experiment, copper ethylenediaminetetraacetate as the flame-retardant component was mixed with the SiO<sub>2</sub> porous particles. Copper ethylenediaminetetraacetate of 17.6 parts by weight was mixed with the porous particles of 100 parts by weight. The pellets of 90wt% and the mixture of copper ethylenediaminetetraacetate and the SiO<sub>2</sub> porous particles of 10wt% were kneaded with the twin screw kneader at 185°C, and press-molded into a test piece of 125mm x 13mm x 3.2mm (at a molding temperature of 180°C under a pressure of 120kg/cm<sup>2</sup>). The SiO<sub>2</sub> porous particles employed in this experiment were the same as those employed in Experiment 1 and finally

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dispersed in the resin in a form of fine particles in the order of nanometer. The content of copper ethylenediaminetetraacetate in the resin composition was calculated to be 1.5wt%.

The resultant test piece was subjected to the UL-94 vertical flame test similarly to Experiment 1. The results are shown in Table 1. From the results shown in Table 1, this sample was evaluated to be V2 according to the UL specification. Ethylenediaminetetraacetate was used in a form of powder which had a particle diameter of about 5 $\mu$ m to about 90 $\mu$ m. The powder was not crushed by kneading and the powder remaining the initial size was dispersed in the resin.

### 7. Results

Item	Experiment 1	Experiment 2	Experiment 3	Experiment 4
Afterflame time	20 sec.	21 sec.	20 sec.	22 sec.
Total afterflame time for 5 samples	120 sec.	125 sec.	130 sec.	131 sec.
Afterflame time after second flame application	26 sec.	25 sec.	26 sec.	25 sec.
Afterflame or afterglow up to holding clamp	No	No	No	No
Cotton indicator ignited by flaming particles or drops	Yes	Yes	Yes	Yes
Rating	V2	V2	V2	V2
Item	Example 1	Example 3	Example 5	Example 8
Afterflame time	11 sec.	11 sec.	9 sec.	13 sec.
Total afterflame time for 5 samples	65 sec.	65 sec.	62 sec.	67 sec.
Afterflame time after second flame application	13 sec.	11 sec.	13 sec.	14 sec.
Afterflame or afterglow up to holding clamp	No	No	No	No
Cotton indicator ignited by flaming particles or drops	No	No	No	No
Rating	V0	V0	V0	V0

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For reference, the results of Examples 1, 3, 5 and 8 shown in Table 3 of the specification of the present application, which are the same as Experiments 1-4 respectively, except that the flame-retardant component are supported by the  $\text{SiO}_2$  porous material. Therefore, the contents of the flame retardant component and the  $\text{SiO}_2$  porous material in the resin composition made in Experiment 1 were the same as those in the resin composition of Example 1. The same can be said for Experiment 2 and Example 3; Experiment 3 and Example 5; and Experiment 4 and Example 8.

As apparent from the table, the resin composition wherein the flame-retardant component is simply mixed with the  $\text{SiO}_2$  porous material (Experiments 1-4) show lower flame-retardancy compared to the resin composition wherein the flame-retardant component is supported by the  $\text{SiO}_2$  porous material (Examples 1, 3, 5 and 8) even if the weight ratio of the  $\text{SiO}_2$  porous material to the flame retardant component, as well as the weight ratio of the flame retardant to the entire resin composition, is the same in both resin compositions made in each Experiment and each Example. This means that the flame retardancy-imparting component supported on the inorganic porous material provides the resin composition with excellent flame retardancy.

8. The undersigned petitioner declares further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true;

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and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of this application or any patent issuing thereon.

Date: August 19, 2008

By Takehiko Yamashita  
Takehiko YAMASHITA